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POLLUTANT TRANSPORT AND ACID RAIN FORMATION IN A FIELD OF CONVECTIVE CLOUDS: RESULTS FROM A MESOSCALE MODEL SIMULATION

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1. INTRODUCTION

Transport, scavenging and chemical transformations of sulphur and nitrogen pollutants in a field of convective clouds are studied in this paper by means of a three-dimensional mesoscale model, with horizontal resolution of 10 km and parameterized subgrid effects. Attention is focused on the average effects of cumulus convection, as well as the relative importance of different microphysical and chemical processes.

The paper is an outcome of the author's stay at the GKSS Forschungszentrum Geesthacht GmbH in Geesthacht, West Germany and the collaboration with its staff on the development of a mesoscale model of transport and transformations of atmospheric pollutants.

The University of Toronto and Ontario Hydro cloud chemistry model (Iribarne and Melo, 1988) has been coupled for this study with the developed at GKSS cloud microphysics module (Levkov et al., 1987) and a parameterization scheme for the subgrid effects of cumulus convection (Levkov et al., 1986a). These models are driven by a relatively simple dynamical model, which will be replaced in future with a sophistcated mesoscale model GESIMA being developed at GKSS. The model is briefly described below. More detailed description is given by Niewiadomski et al. (1989). Detailed formulas can be found in appropriate reports referenced in section 2.

2. THE MODEL

2.1 Dynamics

The dynamical module is based on the model of Beniston (1984). Detailed formulas can be found in papers of Levkov et al. (1986a,b). The model solves the hydrostatically approximated prognostic equations for momentum, potential temperature, specific humidity, hydrometeor mixing ratios, and chemical variables. *The turbulent transport is parameterized through first-order closure; surface fluxes are computed according to Businger et al. (1971). Source terms due to microphysical and chemical processes as well as due to the subgrid effects of cumulus clouds are provided by the described below microphysical, chemical and cloud modules.*

2.2 Cloud microphysics

The model follows the 'bulk microphysics' approach. The liquid water is divided into rain and cloud water fractions. Cloud ice and snow are combined in one wariable, which will be refered to as 'snow'. The following microphysical processes are included: condensation, evaporation and freezing of cloud water, autoconversion of cloud water to rain, sublimational growth of snow, accretion of cloud droplets by rain and snow particles, freezing and evaporation of rain and melting of snow. Transfers of pollutants between cloud water, rain and snow due to microphysical processes are computed in the chemical module. Detailed formulas can be found in Levkov et al. (1987). Various aspects of this model are also discussed by Levkov et al. (1988).

2.3 Chemistry

The chemical module, computing the source terms for chemical variables due to chemical and micro physical processes is a slightly modified version of the model of Iribarne and Melo (1988). Its basic ideas are

described by Iribarne and Cho (1988). The rates of the microphysical transfer processes are provided by the microphysical module.

The following chemical processes are included in the model:

a. Oxidation of SO_2 in cloud and rain water by H_2O_2 dissolved from the air.

b. Oxidation of SO_2 in cloud and rain water by O_3 dissolved from the air.

c. Oxidation of NO_2 by OH in gaseous phase with subsequent dissolution in cloud water of the HNO_3 formed.

d. Oxidation of NO by OH in gaseous phase with subsequent dissolution in cloud water of the HNO_2 formed.

Note, that while the H_2O_2 concentration in both, cloud and rain water is a separate prognostic variable, the air concentrations of ozone and OH, as well as CO_2, are assumed constant. No chemical reactions take place in the ice phase, the snow is however a medium of vertical transport of pollutants and of their transfer between cloud water and rain.

2.4 Cumulus parameterization

Parameterization of the subgrid effects of cumulus convection is based on the ideas of Augstein and Wendel (1981). The scheme, which uses a simple, one-dimensional, entraining cumulus model to obtain the cloud parameters is briefly described below. For details see Levkov et al. (1986a).

The scheme is applied in each column of the model, where the large scale vertical velocity (computed by the dynamical module) exceeds a prescribed value at the lifting condensation level. If that criterion is fulfilled, the in-cloud parameters (temperature, momentum, hydrometeor mixing ratios, and chemical variables) are computed with the 1-D cloud model, using the microphysical parameterization described above, and the entrainment coefficient given by an exponentially increasing function of the altitude above the cloud base (LCL). The model is started at the cloud base with vertical velocity equal to the Deardorff's convective velocity scale. The computed vertical velocity is at each level reduced by a constant factor accounting for the partial coverage of the grid cell by clouds.

For each prognostic variable of the model, effects of convection are then assumed at each level as proportional to the in-cloud vertical velocity, entrainment function and the difference between the values of the considered variable in and outside the cloud (i.e. between the values provided by the dynamical module and the 1-D cloud model).

3. THE EXPERIMENT

The model was run with time steps of 20 s in a domain of 200 x 200 x 10 km with horizontal resolution of 10 km and vertical grid step 50 m for the lowest layer and 500 m for all other ones. Three runs of the model, with two pollution scenarios, imposed on two meteorological cases with different characteristics of microphysical processes, were performed. Their results are discussed in section 4. The input data for these cases are described below. The model was run for two hours of real time, simulating therefore rather early stages of the cloud development. For each run this time was however significant precipitation reaching the ground.

The meteorological conditions used in the runs 1 and 3 represent moderate latitude maritime convection, and are derived from the 13 Oct. 1981 case of the North Sea KonTur experiment. The data for the run 2 come from the 2 Sept. 1974 case of GATE and is characteristic for the intertropical convergence zone.

The behaviour of the dynamical and microphysical parts of the model for these two cases is discussed by Levkov et al. (1986a, 1986b). The rainfall rate, as well as the cloud water and snow mixing ratio at the

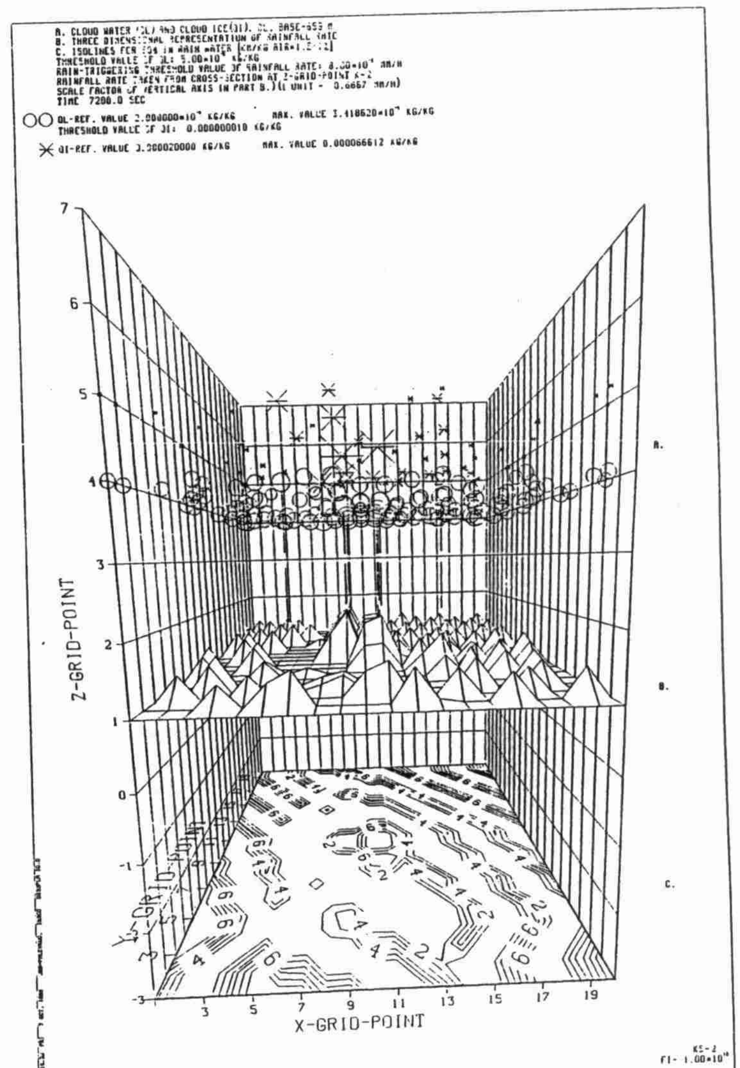

Fig. 1.

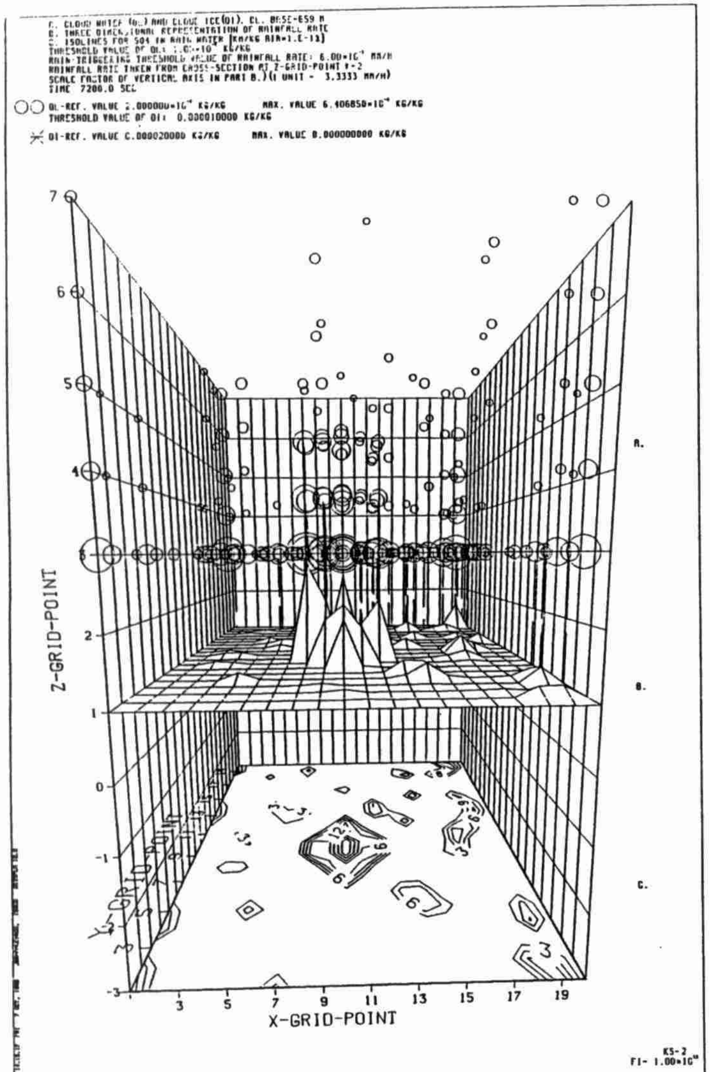

Fig. 2.

end of simulations, are presented at Fig.1 (run 1) and Fig.2 (run 2), along with isolines of rainwater sulphate content close to the ground. The tropical case (run 2) produces much more cloud water and, eventually more rain, but the rain develops slower and by the end of the second hour of simulation its concentration is in some grid cells higher, but the total content in the domain lower than in the KonTur case (runs 1 and 3).

The two arbitrarily assumed pollution cases represent summer (runs 1 and 2) and winter (run 3) conditions of moderate pollution. For each chemical variable of the model a uniform concentration was assumed in the lowest levels of the model (up to 1500 m) and another, uniform concentration above that level. These initial profiles are presented in Table 1. The main difference between these cases is the much lower concentration of H_2O_2 for the winter case.

TABLE 1.

Initial concentrations, a: below 1500 m, b: above 1500 m

		run 1&2 (a)	run 1&2 (b)	run 3 (a)	run 3 (b)
SO_2	(ppb)	15	0.10	15	0.10
NO	(ppb)	1	0.02	5	0.02
NO_2	(ppb)	4	0.08	25	0.08
n_A	(10^{-11} kg eq/ kg air)	3	3	3	3
$SO_4 =$	(10^{-9} kmole/ kg air)	2	0.2	2	0.2
H_2O_2	(ppb)	1	2	0.1	0.2
O_3	(ppb)	100	70	30	30
OH	(10^{-14} atm)	5	5	0.5	0.5

4. RESULTS

Elements of the sulphate budget are presented in Table 2. The values given in that table are, the integrated over the whole domain and two hours of simulation, source terms for the sulphate variables, computed in the chemical module. The difference between the source and sink terms is the contribution of in-cloud and below-cloud processes to the total content of sulphate in the domain of the model. No snow melting took place in any case.

As can be seen, the hydrogen peroxide is the main oxidant. Even in the run 3, with low H_2O_2 content, more SO_2 is oxidized by H_2O_2 then by ozone. The below cloud scavenging was of secondary importance, especially in summer cases. Comparison of the results of the runs 1 and 3, with the same microphysics (i.e. nucleation scavenging) but different oxidant content, shows that in the summer case the oxidation accounts for roughly 50% of the deposited sulphate, the remaining part coming from the nucleation and below-cloud scavenging of aerosol. The effects of oxidation were, even for run 3, significantly higher then those obtained by Niewiadomski (1989) for a field of Cu clouds, with a model with resolution of 1 km and H_2O_2 reaction removed. This difference results, at least partly, from the lower coverage by clouds in that model.

During the two hours of simulation the oxidation consumed about 2% (run 1), 7% (run 2), and 0.4% (run 3) of the SO_2 available in the domain. The consumption of H_2O_2 was 4%, 14%, and 4% respectively.

The oxidation of NO and NO_2 accounted for about 50% of nA (acidity parameter) produced in the model in run 1, 25% in run 2 and 79% in run 3. However only a fraction of the nA produced in gas phase by NOx oxidation can actually enter the cloud water and affect the acidity of rain. Rain deposited 146 kg equiv of nitrate in run 1, 76 in run 2 and 93 in run 3.

5. CONCLUSIONS

The chemical, microphysical and cumulus parameterization modules coupled for this study proved to be an effective tool for studying the mesoscale effects of cumulus clouds. After further improvements the modules will be incorporated in a more sophisticated dynamical model.

H_2O_2 was the main oxidant in cases studied, with sulphate production strongly dependent on the ammount of H_2O_2 available. Sulphate production was depended also on the ammount of liquid water in the domain. NOx oxidation was of secondary importance in this study.

The average vertical profiles of SO_2 in air and cloud water are given Fig.3 for run 1 in 30 min intervals. The upward transport of SO_2 is clearly seen. The profiles resemble those obtained by Niewiadomski (1966) for a passive pollutant with a resolution of 1 km.

TABLE 2.

The sulphate budget, kg equivalents.

			run 1	run 2	run 3
Produced by SO_2 oxidation:					
in cloud water,	by	H_2O_2	1510	5100	147
in cloud water,	by	O_3	50	20	117
in rain,	by	H_2O_2	7.9	0.9	2.6
in rain,	by	O_3	0.2	0.5	0.9
cloud to rain transfer			598	851	237
cloud to snow transfer			23	0	8.5
below cloud scavenging			144	44	129
wet deposition			578	220	280

6. ACKNOWLEDGEMENT

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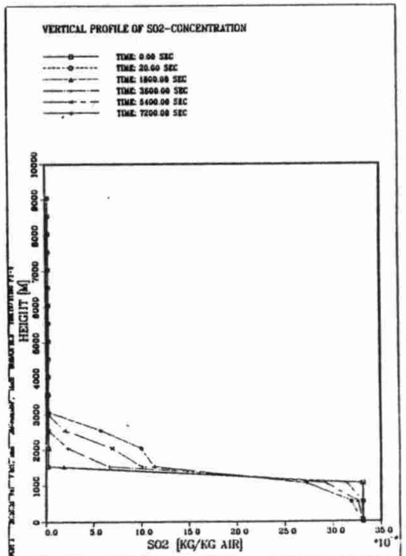

Fig. 3